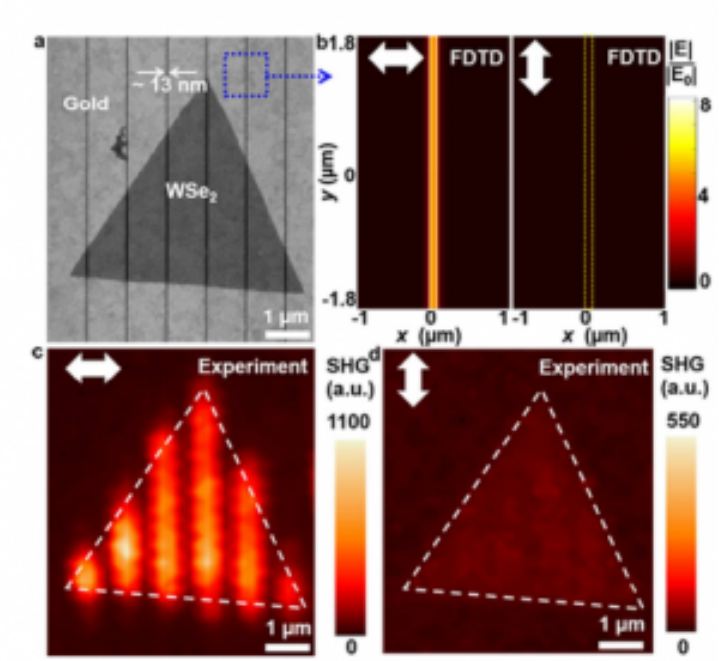
Selectively Plasmon-Enhanced Second-Harmonic Generation from Monolayer Tungsten Diselenide on Flexible Substrates



. Pump-laser-polarization dependent SHG mapping. (a) SEM image of single-crystalline monolayer WSe2 flake on trenches with a pitch of 910 nm. (b) Simulated electric field distribution at a plane 1 nm above the surface of gold substrate with pump laser polarized perpendicular (left panel) and parallel (right panel) to the trench. The dotted line outlines the geometry of the trench. (c,d) Corresponding experimental SHG mappings of the exact WSe2 flake on trenches as shown in the SEM image in (a) under resonant and non-resonant excitations, respectively. White dashed lines outline the WSe2 flake. The white arrows show the polarization directions of the pump laser.

Monolayer two-dimensional transition metal dichalcogenides (2D TMDCs) exhibit promising characteristics in miniaturized nonlinear optical frequency converters, due to their inversion asymmetry and large second-order nonlinear susceptibility. However, these materials usually have a very short light interaction lengths with the pump laser because they are atomically thin, such that second-harmonic generation (SHG) is generally inefficient. In this research, Joel.K.W.Yangs group fabricated a judiciously structured 150-nm-thick planar surface consisting of monolayer tungsten diselenide and sub-20-nm-wide gold trenches on flexible substrates, reporting ~7000-fold SHG enhancement without peak broadening or background in the spectra as compared to WSe2 on as-grown sapphire substrates. their proof-of-concept experiment yields effective second-order nonlinear susceptibility of 2.1 × 104 pm/V. Three orders of magnitude enhancement is maintained with pump wavelength ranging from 800 nm to 900 nm, breaking the limitation of narrow pump wavelength range for cavity-enhanced SHG. In addition, SHG amplitude can be dynamically controlled via selective excitation of the lateral gap plasmon by rotating the laser polarization. Such fully open, flat and ultrathin profile enables a great variety of functional samples with high SHG from one patterned silicon substrate, favoring scalable production of nonlinear converters. The surface accessibility also enables integration with other optical components for information processing in an ultrathin and flexible form.